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PERFORMANCE CHARACTERIZATION OF A BOSCH CO₂ REDUCTION SUBSYSTEM

FINAL REPORT

by

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February, 1980

by

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for

AMES RESEARCH CENTER
National Aeronautics and Space Administration

FOREWORD

The development work described herein was conducted by Life Systems, Inc. for the National Aeronautics and Space Administration (NASA) Ames Research Center. The period of performance for the program was June, 1979 to January, 1980. The Program Manager was Dennis B. Heppner, Ph.D. Support was provided by Tim M. Hallick in the program testing, data reduction and analysis and Franz H. Schubert in the test design and data analysis.

The contract's Technical Monitor was P. D. Quattrone, Chief, Advanced Life Support Office, NASA Ames Research Center, Moffett Field, CA.

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LIST OF ACRONYMS

ARS	Air Revitalization System
B-CRS	Bosch Carbon Dioxide Reduction Subsystem
EDC	Electrochemical Depolarized CO_2 Concentrator
OGS	Oxygen Generation Subsystem
ORS	Oxygen Recovery System
RLSE	Regenerative Life Support Evaluation
TSA	Test Support Accessories

SUMMARY

Regenerative processes for the revitalization of spacecraft atmospheres are
needed to sustain man in space for extended periods of time. ·A major step in
this revitalization process is the recovery of oxygen, i.e., the collection of
metabolic carbon dioxide and water vapor and the recovery of oxygen from these
products. One uniquely qualified component for the overall oxygen recovery
process is the Bosch Carbon Dioxide Reduction Subsystem. In the recent past,
major emphasis has been placed on the development of a Carbon Dioxide Reduc-
tion Subsystem based on the Sabatier process. However, the development of the
alternate Bosch carbon dioxide reduction technique is required to answer the
needs that will result from a Shuttle or spaceflight imposed requirement to
minimize or eliminate the overboard venting of gases and/or to reduce equiva-
lent launch weight for extended duration missions. The Sabatier reaction
products are gaseous while the Bosch product is solid carbon, which can be
stored on-board. Also, oxygen is lost overboard in the Sabatier reactor
exhaust in the form of unreacted carbon dioxide.

The Bosch Carbon Dioxide Reduction Subsystem has been under study for many
years, and various versions of hardware have been developed. The objective of
a majority of prior programs had been to answer basic hardware questions such
as how to develop working reactors and what materials and components are
needed to support the reactors. Much less attention had been given to charac-
terization of Bosch performance at the subsystem level to derive operating
conditions that result in minimum weight, volume and power.

The study reported herein is the performance characterization of Bosch hardware
at the subsystem level (up to five-person capacity) in terms of five operating
parameters. Selection of the five operating parameters was based on Life
Systems' prior operating experience with Bosch hardware and the importance of
these parameters, as cited in other work. The five parameters were: (1)
reactor temperature, (2) recycle loop mass flow rate, (3) recycle loop gas
composition (% hydrogen), (4) recycle loop dew point and (5) catalyst density.
Experiments were designed and conducted in which the five operating parameters
were varied and Bosch performance recorded. A total of 12 carbon collection
cartridges provided over approximately 250 hours of operating time. Generally,
one cartridge was used for each parameter that was varied.

The testing program was successfully completed. The Bosch hardware performed
reliably and reproducibly. No startup, reaction initiation or carbon contain-
ment problems were observed. Optimum performance points/ranges were identified
for the five parameters investigated. The performance curves agreed with
theoretical projections. The results provide needed information for future
Bosch subsystem designs.

INTRODUCTION

Regenerative processes for the revitalization of spacecraft atmospheres are
essential in making long-term manned missions in space a reality.[1]   An
important step in this overall revitalization process is the collection and

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(1) References cited at end of report.

concentration of the metabolically-produced carbon dioxide (CO_2) for breathing oxygen (O_2) recovery. Stripping the O_2 from the CO_2 is performed in the subsequent step of CO_2 reduction. The Bosch CO_2 Reduction Subsystem (B-CRS) performs this function. The subject program continued the development of this subsystem by further characterizing the Bosch process.

Background

Providing O_2 to the cabin atmosphere of a manned spacecraft is an important function of a regenerative Environmental Control Life Support System (EC/LSS). Within an EC/LSS, various subsystems of the Air Revitalization System (ARS) are responsible for the regeneration of metabolically-consumed O_2 and the removal and concentration of the metabolically-generated CO_2 from the atmosphere. For long-duration or large crew manned space missions, launch and supply requirements can be materially reduced and simplified by in-flight reclamation of O_2 from concentrated CO_2. The Space Shuttle era provides both an opportunity for extensive testing of such regenerative systems and marks the beginning of the time period in which they will be required.

Three life support subsystems are uniquely qualified to perform the function of reclaiming O_2 within an ARS. They are an Electrochemical Depolarized CO_2 Concentrator (EDC),[2,3] an O_2 Generation Subsystem (OGS)[4] and a B-CRS. Figure 1 is a simplified block diagram of a closed O_2 loop which integrates these three subsystems with the spacecraft's atmosphere. Oxygen and hydrogen (H_2) are generated through the electrolysis of water by the OGS. Carbon dioxide is stripped from the cabin atmosphere by the EDC and is sent mixed with H_2 to the B-CRS. The B-CRS reduces the CO_2 to form carbon and water. The water is returned to the OGS for subsequent regeneration of the O_2 and H_2.

Grouping the three subsystems into an integrated O_2 Recovery System (ORS) results in total recovery of the O_2 from metabolically-generated CO_2, i.e., permitting closure of the metabolic O_2 loop. This factor offers an advantage over the Sabatier-based CO_2 reduction concept where O_2, in the form of unreacted CO_2, is vented overboard. This loss in O_2 implies increased storage of O_2 or water (electrolysis), resulting in an increase in spacecraft launch weight. Another significant advantage is that the Bosch-based ORS can eliminate the overboard dump requirement for gases which may contaminate the immediate spacecraft environment. Continued technology development is required to establish the Bosch as an alternate CO_2 reduction process.

State-of-the-art technology and hardware for the B-CRS has been advanced under the cognizance of NASA's Marshall Space Flight Center (MSFC) and Ames Research Center (ARC). Over the past eight years, MSFC has developed various versions of Bosch reactors.[5-7] These reactor development programs investigated materials and techniques for reactor design and demonstrated reliable Bosch operation over 15,000 hours of testing. Ames Research Center has supported[8-11] studies of the Bosch process at the fundamental chemical reaction level. Over 3,000 hours of testing in these studies have provided a good understanding of the process reaction dynamics.

Since 1975, Life Systems, under MSFC sponsorship and supported by ARC, has developed hardware for and conducted tests of the integration of the Bosch with the EDC[12,13] and of the combined B-CRS/EDC with an OGS.[14] Over 3,300
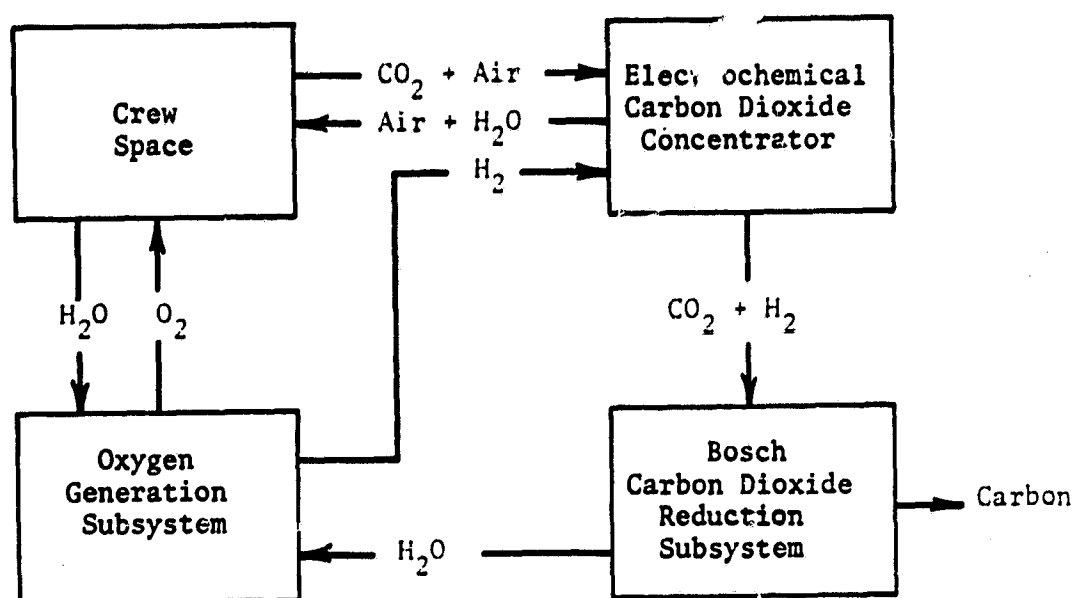
FIGURE 1   CLOSED-LOOP OXYGEN RECOVERY SYSTEM

hours of Bosch operation has been logged at Life Systems. To date, the integration and development tests have removed all technical and operational problems involved with introducing the B-CRS into an ORS. The testing results demonstrated the B-CRS is ready for preprototype development.[15] The activities performed under the current program obtained performance data at the subsystem level that is needed to design a preprototype B-CRS.

Program Objectives

The program's primary objective was to obtain, through actual testing, the engineering parametric data required to optimize the design of the next generation of B-CRS for spacecraft application. Achieving this objective required operating an existing B-CRS and determining its performance in response to variations of several key operating parameters. The selected parameters and experiment protocol were generalized to minimize the effect of the specific test hardware as much as possible. The test program was designed to simulate projected spacecraft requirements in terms of how the B-CRS might interface with other EC/LSS elements.

Program Organization

To meet the above objective, the program was divided into three tasks plus the documentation and program management functions. These three tasks were:

1. Modify the existing B-CRS Test Facility to accommodate test program requirements by adding control and measurement capability and modifying the primary H_2 and CO_2 gas feed hardware.

2. Perform tests to determine the effects of five key operating parameters on B-CRS performance, i.e., CO_2 reduction rate: (1) reactor operating temperature, (2) recycle loop composition (% H_2), (3) recycle loop mass flow rate, (4) recycle loop dew point and (5) catalyst density.

3. Prepare a report documenting the results of the testing program.

Report Organization

This Final Report covers the work performed from June, 1979 through January, 1980. The following three sections present the technical results grouped according to (1) Bosch CO_2 Reduction Process, (2) B-CRS Test Facility and (3) Program Testing. These sections are followed by Conclusions and Recommendations based on the work performed.

BOSCH CO_2 REDUCTION

Carbon dioxide can be reduced to solid carbon and water by reaction with H_2. Detailed descriptions of CO_2 reduction using the Bosch process, its theory of operation, specific hardware developments and performance have been discussed previously.[5,8,12] The following summarizes the Bosch reactions, process, hardware and operation.

Reactions and Process Description

The Bosch reaction occurs in the range 800 to 1000 K (980 to 1340 F) in the presence of an iron catalyst. Carbon dioxide combines with H_2 and produces carbon and water vapor as indicated in the overall reaction:

$$CO_2 + 2 H_2 = C + 2 H_2O + Heat \tag{1}$$
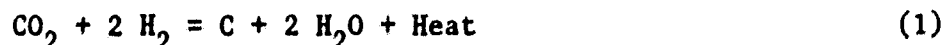
$$Heat = 2.26 \ MJ/kg \ CO_2 \ (975 \ BTU/lb \ CO_2)$$

One mole of CO_2 combines with two moles of H_2 to form one mole of carbon and two moles of water vapor. In practice, single pass efficiencies through the Bosch reactor are less than 10%. Complete conversion is obtained by recycling the process gases with continuous deposition of carbon and removal of water vapor. The recycled gas mixture contains CO_2, H_2, water vapor, carbon monoxide (CO) and methane (CH_4). These components are formed by intermediate reactions, such as:

$$CO_2 + C = 2 \ CO \tag{2}$$

$$CO_2 + H_2 = CO + H_2O \tag{3}$$

$$CO + H_2 = C + H_2O \tag{4}$$

$$2 \ H_2 + C = CH_4 \tag{5}$$

An equilibrium condition for the gas mixture is reached based on the specific operating temperatures, pressures and relative proportions of the primary reactants, CO_2 and H_2.

Simplified Schematic and Operation

Figure 2 is a simplified flow schematic of a B-CRS identifying the major subsystem components. The B-CRS operates as follows. Gases are continuously circulated through the recycle loop by a compressor. The gases leaving the compressor are diverted by two ganged valves to either of the two regenerative heat exchanger/reactor combinations. The gases are preheated in the respective regenerative heat exchanger prior to entering the reactor. Within the reactor CO_2 and H_2 react over an iron catalyst in the volumetric ratio of 2:1 (H_2:CO_2) to form carbon and water vapor. The recycle gases, partially depleted in CO_2 and H_2 leave the reactor at a temperature near 922 K (1200 F) to exchange heat with the incoming gases in the regenerative heat exchanger. The mixture then flows through the valves to a condenser/separator where the water vapor is condensed, separated and collected. The recycle loop gas mixture then returns to the compressor.

The feed gases (H_2 and CO_2) are added to the loop upstream of the compressor. This allows the feed gas pressure to remain at a minimum. For practical applications the ratio of recycled gas flow rate to feed gas flow rate is in the range of about 15-20 to 1, indicating that conversion efficiency per pass through to reactor is about 6%.

Process rate control is required due to changes in the amounts of feed gases entering the recycle loop, reaction rate changes, pressure changes, composi-
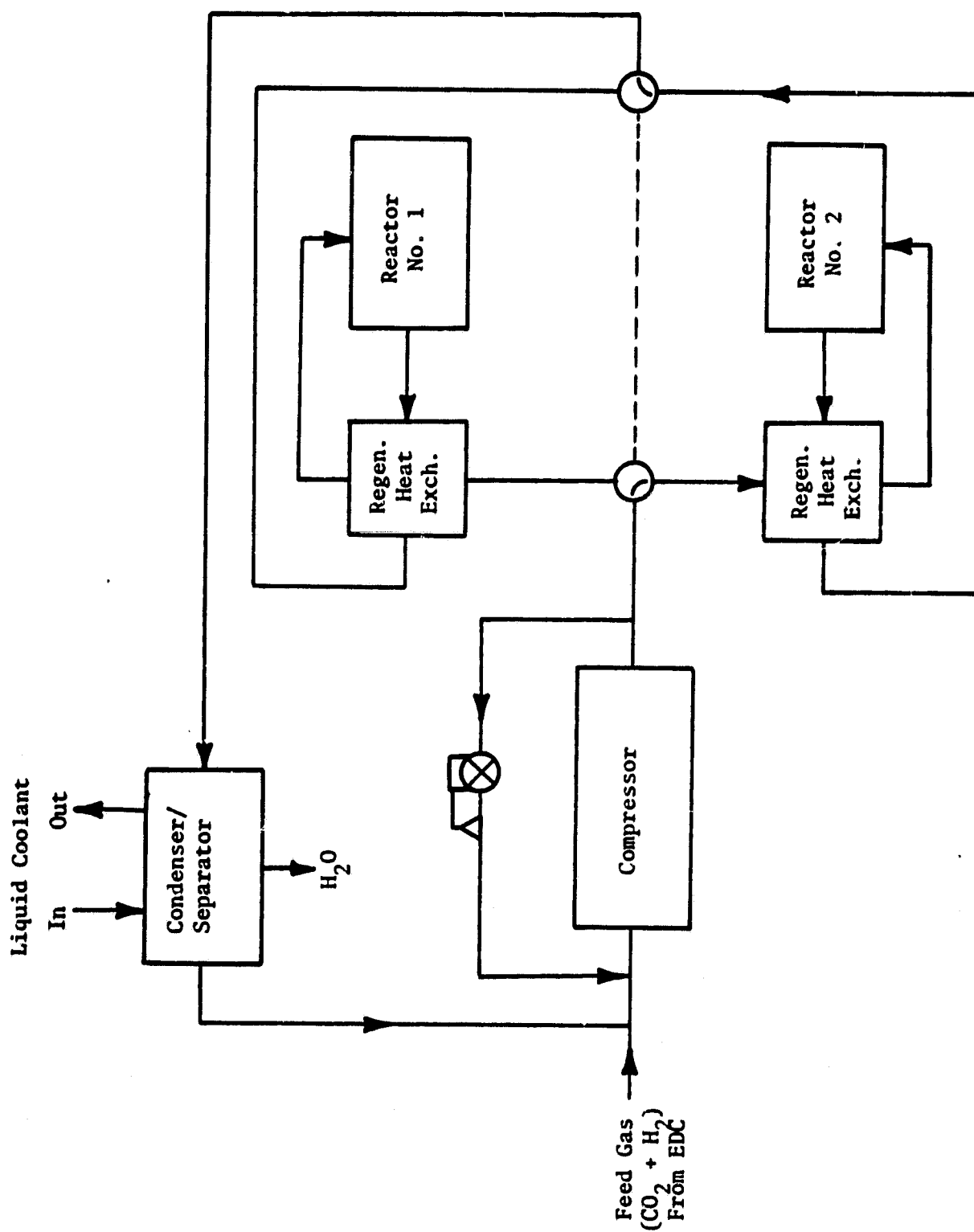
FIGURE 2 SIMPLIFIED B-CRS FLOW SCHEMATIC

tion changes, etc. A pressure regulator bypassing recycle flow from the constant speed compressor achieves this process rate control by maintaining a constant feed pressure. As conversion efficiency drops or feed gas rate increases, the overall pressure level in the recycle loop tends to rise. This is resisted by the closing of the pressure regulator, thus increasing the recycle flow through the reactor; the feed pressure will then be reduced as the reaction efficiency is increased and more gases are consumed. Conversely, a decrease in recycle loop flow will result from a decrease in feed gas flow rate and/or an increase in process reaction efficiency. This technique effectively maintains dynamic rate control for small pertubations in process response.

The carbon remains in the reactor and is collected in expendable cartridges. A dual reactor system is used to enable continuous operation. This allows collection of carbon in one reactor while the other remains dormant until the first reactor is filled with carbon. A typical carbon collection cartridge is shown in Figure 3 with a disassembled view shown in Figure 4. It contains the iron catalyst in the form of common steel wool. The reaction initially occurs on the steel wool surface but later on can occur at previously deposited carbon sites. The total catalyst surface area can therefore be far less than the surface area of the deposited carbon. An expended cartridge filled with carbon is shown in Figure 5. The cartridge concept retains the carbon in a compact, easily handled form.

Major Operating Parameters

The major operating parameters affecting process rates and CO_2 reduction efficiencies are:

1. Flow rate of the recycle gases through the reactor
2. Physical and chemical characteristics of the catalyst
3. Operating temperature of the reactor
4. Operating pressure
5. Composition of the recycle loop gases

Combining thermodynamic and chemical theory with empirical data indicates that these parameters are related in the following manner:

1. Increasing the recycle flow rate increases the process rate (conversion of CO_2 and H_2 into carbon and water).

2. Increasing catalyst surface area increases reaction rate and decreases startup time. Pretreatment of the catalyst also increases reaction rate and decreases reaction startup times.

3. Operating at the higher end of the desired temperature range of 800 to 1000 K (980 to 1340 F) increases the process rate.

4. Changing gas pressures have only minor effects on reaction rate. However, increasing system pressure improves condenser performance and results in a lower content of water vapor in the recycle loop. Increased pressure, on the other hand, complicates component design, especially for high temperature components, and complicates the interfaces with other subsystems.

7

Recycle Loop Gases Inlet and Heater Well

Thermocouple Well

Reactor Cover
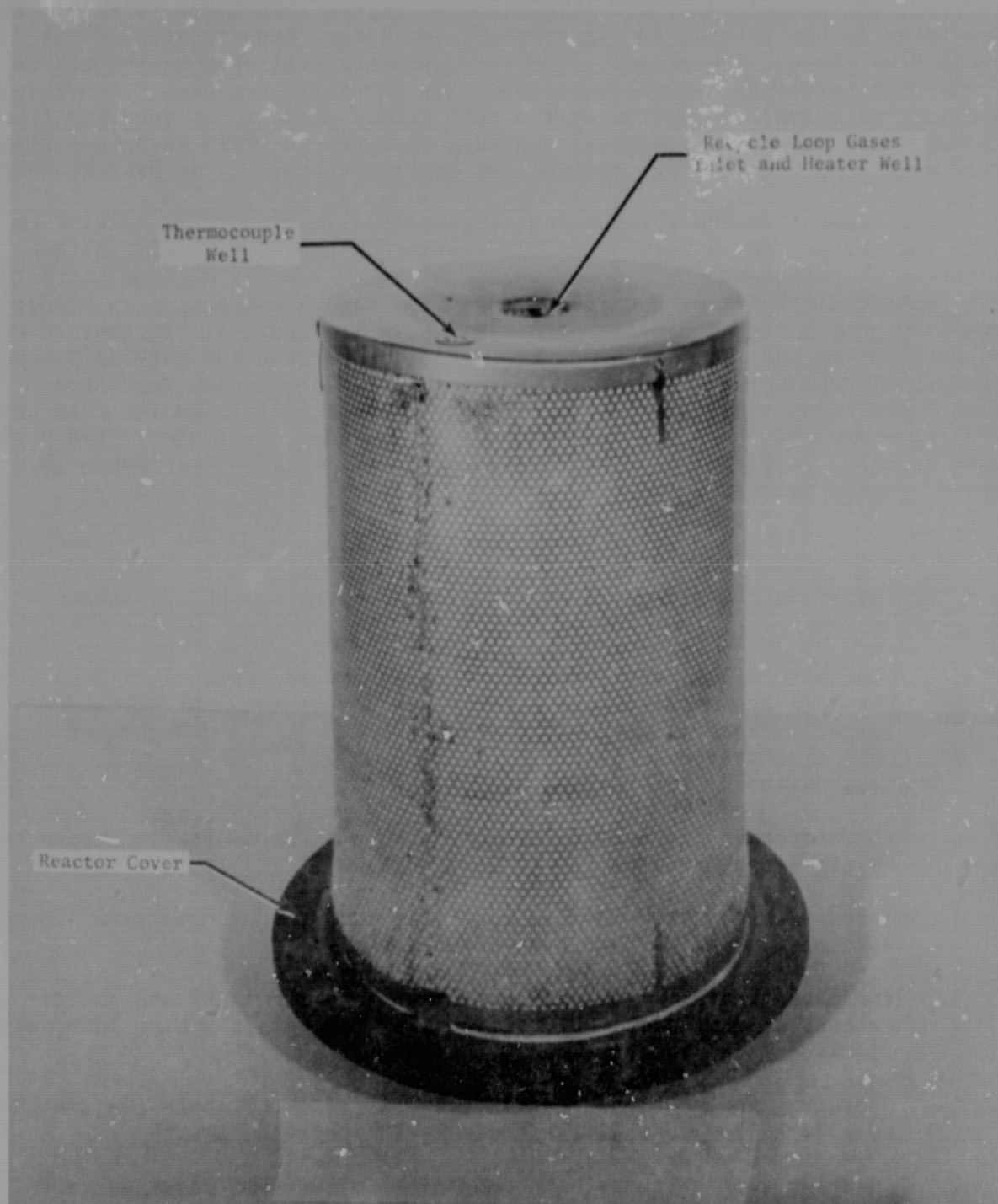
FIGURE 3 CARBON COLLECTION CARTRIDGE (ASSEMBLED)

8

Cartridge Shell
with Fiber Blanket

Cartridge Center Post
with Fiber Blanket

Reactor
Thermocouple Well

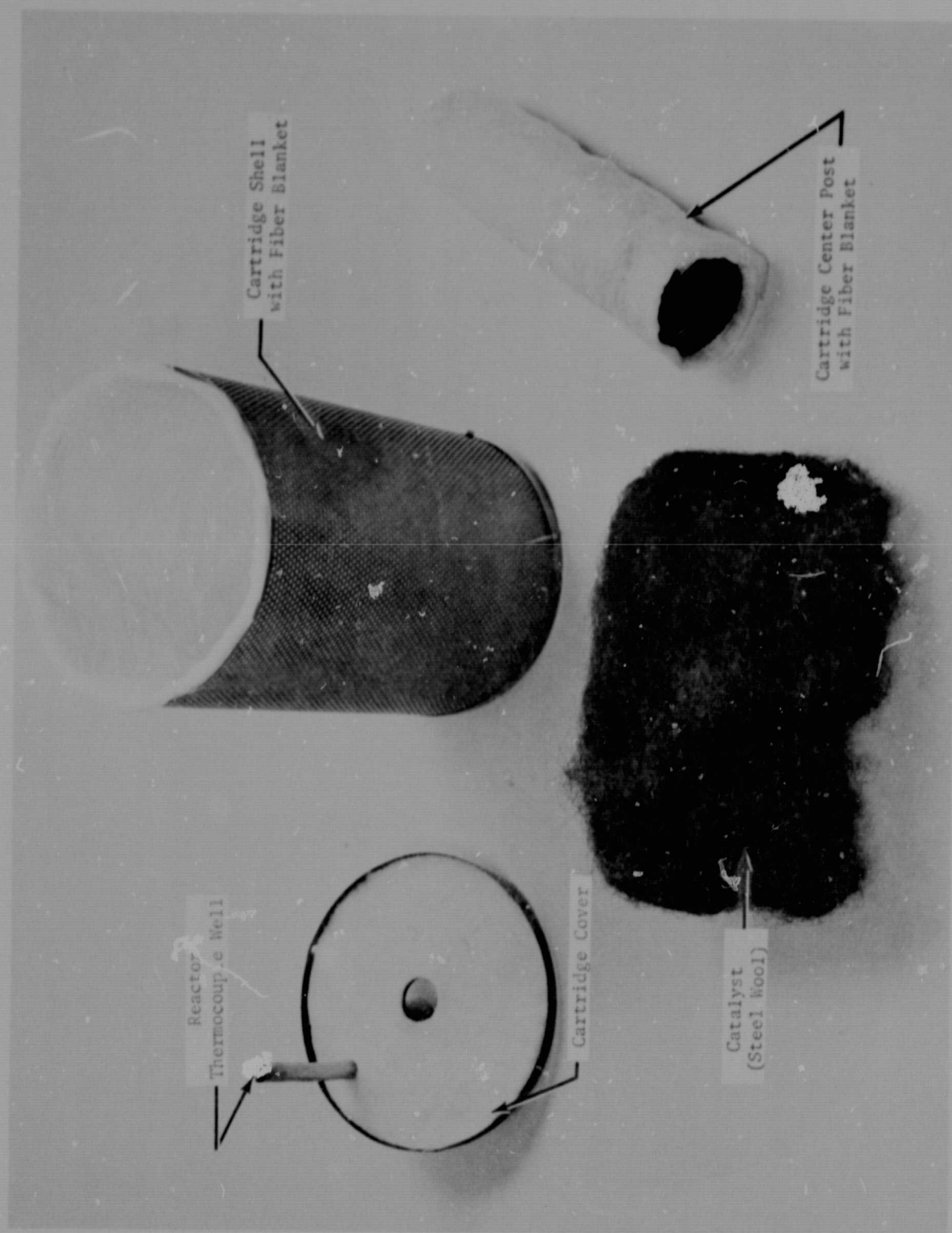Cartridge Cover

Catalyst
(Steel Wool)

FIGURE 4 CARBON COLLECTION CARTRIDGE (DISASSEMBLED)

FIGURE 5 COLLECTION CARTRIDGE FILLED WITH CARBON

5. Changing the composition of the recycle loop gases affects process rate; for example, increasing water vapor partial pressure in the recycle loop will decrease the reaction rate. In general, the composition will reach an equilibrium that corresponds to a given set of subsystem temperatures, pressures, catalyst configuration and flow rates.

The above qualitative observations have been accumulated over many hours of testing. The objective of the current program was to quantify the influence of these factors on CO_2 reduction performance for a given B-CRS hardware. Extrapolation to other hardware designs can then be done through engineering analysis.

B-CRS TEST FACILITY

Life Systems' B-CRS Test Facility was developed principally to support integrated testing of the Bosch with an EDC and OGS.[14] However, the B-CRS can be operated as an independent subsystem as was done for this program. The following subsections discuss the hardware, the subsystem schematic and operation and aspects of the Test Support Accessories (TSA) that were developed or modified for testing.

Hardware Description

Figure 6 shows the B-CRS Test Facility emphasizing the dual-reactor B-CRS and its TSA. The B-CRS consists of two independent reactors, each with its own set of controls and sensors. The B-CRS reactors are nominally sized at the four-person level, meaning they will reduce CO_2 at the rate of 4.0 kg/d (8.8 lb/d). Actual operation shows the maximum CO_2 reduction rate can be somewhat greater than that needed for five persons. Table 1 lists the design operating conditions corresponding to the nominal four-person operation. The carbon collection cartridges are sized for 12 person-days.

Subsystem Schematic and Operation

The mechanical B-CRS consists of the reactors/heat exchangers, compressor, condenser/separator, valving, sensors, ancillary components, plumbing and frame. A schematic of the B-CRS with the TSA is given in Figure 7. To simulate the interface with an EDC, a mixture of H_2 and CO_2 (40% CO_2) is supplied from bottles to the recycle loop through valves V3 and MV15. Feed flow rate and composition are monitored by F1 and A2 respectively. Since the feed mixture is lean in H_2 ($H_2:CO_2$ is 1.5:1 versus 2:1 stoichiometric), additional H_2 enters the loop through valves MV17 and V5 and flowmeter FM3. The added amount is controlled by the loop composition controller (A1) which maintains the recycle loop H_2 mole fraction to the desired setpoint. The total volumetric flow ratio of feed H_2 to CO_2 at any one time, neglecting small variations, is two to one. The CO_2 supply provides for initial purging through MV3 and prevention of a subatmospheric condition in the reactor during cool-down by admitting makeup gas through V4.

The recycle flow is established by the diaphram compressor (M1) and the recycle flow control valve MV5. Selection of the operational reactor is by MV8 which switches both inlet and outlet streams. Only reactor No. 1 was used during these tests in order to eliminate any differences between reactors affecting

Test Support Accessories
Control Unit

Bosch Test
Support Accessories

Interface Hardware
and Controls

Bosch CO$_2$ Reduction
Subsystem (BRS)

Reactor No. 1

Reactor No. 2
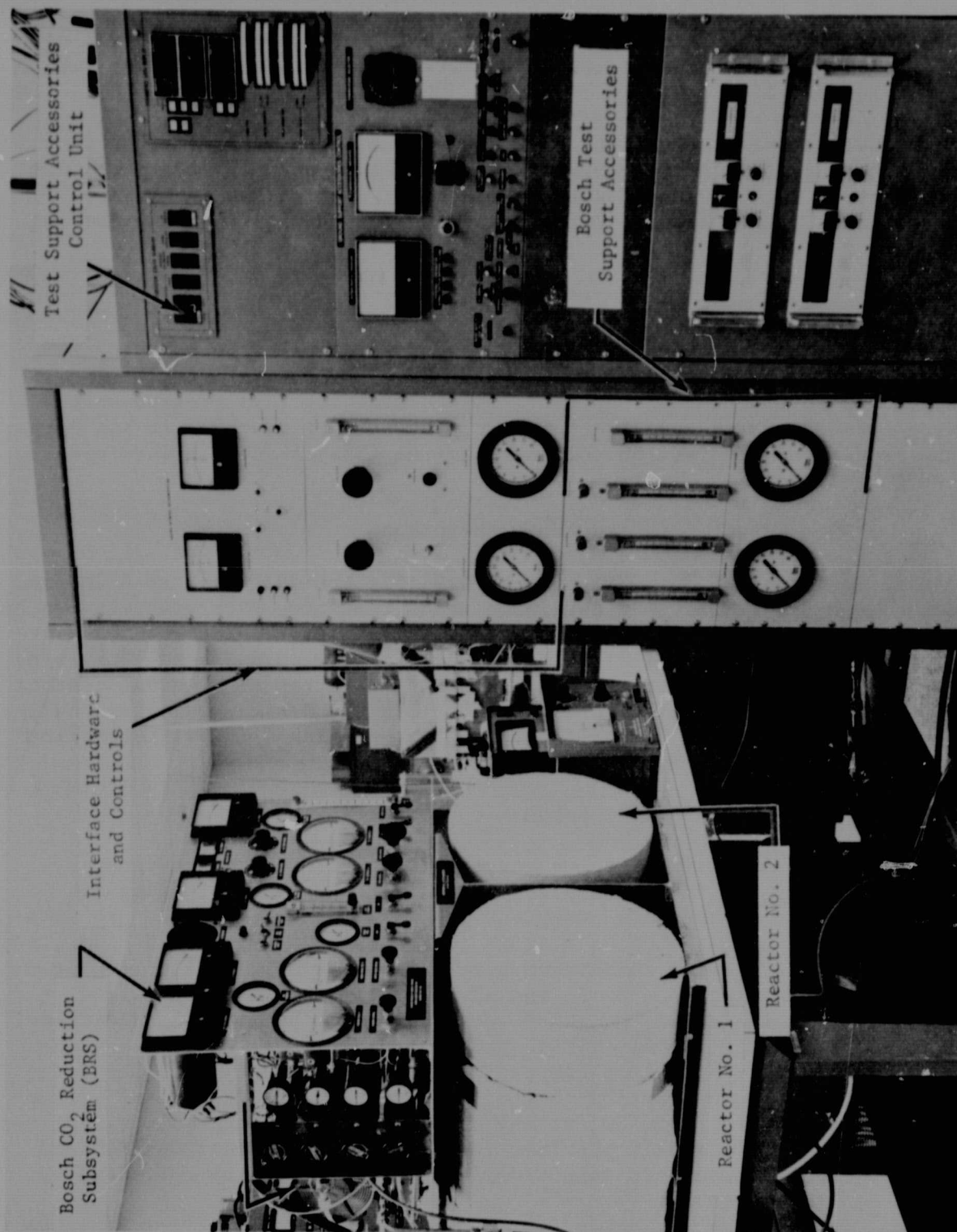
FIGURE 6 B-CRS TEST FACILITY

TABLE 1 B-CRS DESIGN OPERATING CONDITIONS

Feed Gas

CO_2 Flow Rate, kg/h (lb/h)	0.167 (0.367)
H_2 Flow Rate, kg/h (lb/h)	0.0152 (0.0334)
Mixture Ratios	
H_2 to CO_2 by Volume	2
CO_2 to H_2 by Weight	11

Recycle Loop

Flow Rate, kg/h (lb/h)	3.1 (6.8)
Composition, Reactor Inlet, Mole %	
H_2	32
CO	27
CH_4	23
CO_2	16
Water	2
Composition, Reactor Exhaust, Mole %	
H_2	28
CO	28
CH_4	23
CO_2	14
Water	7
Feed Point Pressure, kPa (psia)	105 (15.3)
Gas Temperature, K (F)	
Reactor Out	920 (1196)
Condenser Out	290 (62)
Compressor Out	320 (116)

Condenser/Separator

Coolant	
Type	Water/Antifreeze
Flow Rate, kg/h (lb/h)	60 (130)
Supply Temperature, K (F)	276 (38)
Condensate, kg/h (lb/h)	0.136 (0.300)

Carbon Produced, kg/h (lb/h)   0.045 (0.100)

FIGURE 7   B-CRS TEST FACILITY SCHEMATIC

results. Flow rate to the reactor is sensed by a laminar flow element and differential pressure gauge (PG5). The bypass control (MV7) and bypass return (MV9) are used when changing reactors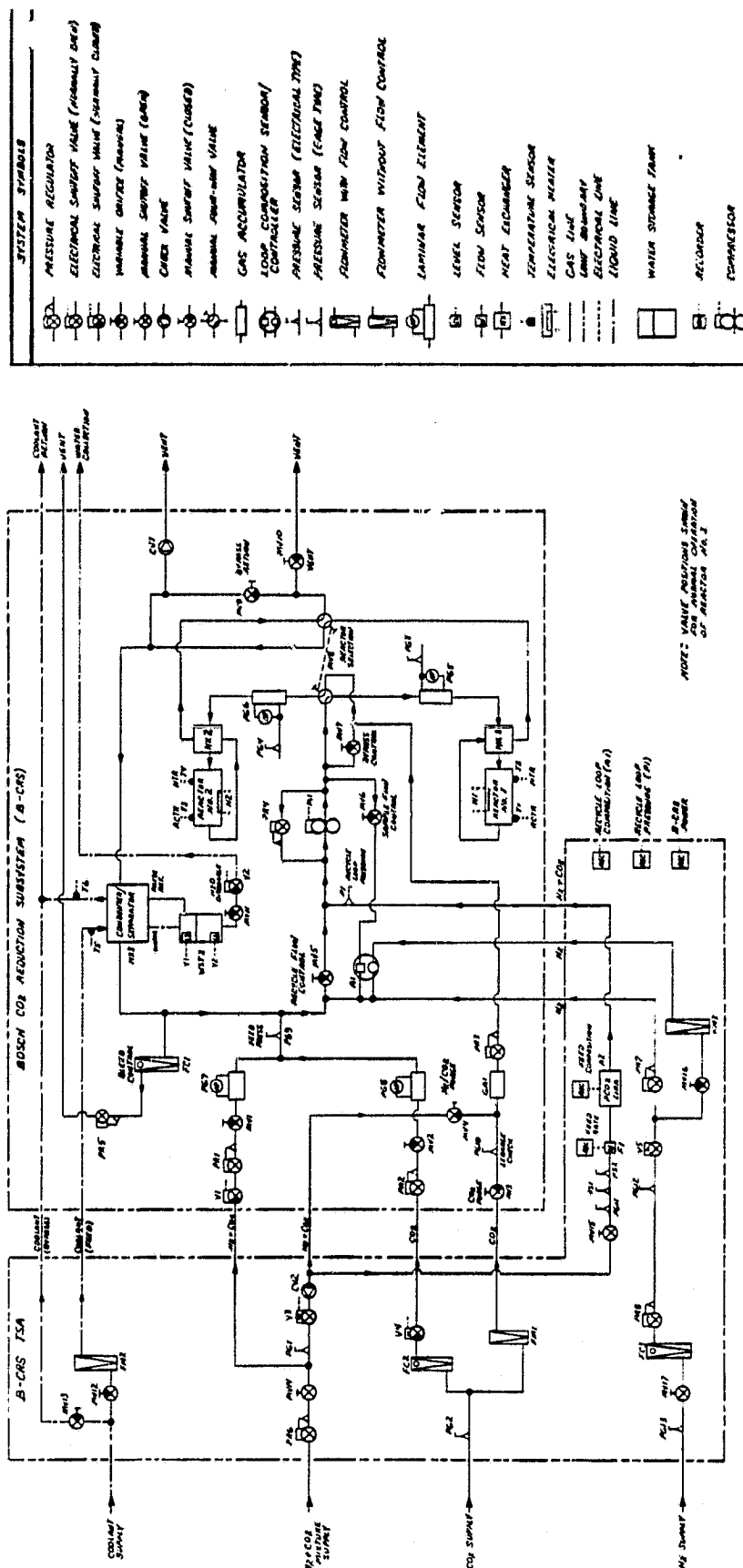 and were not operated during this testing. Following exit of the reactor, the recycle gases flow through the condenser/ separator and back to the compressor.

Coolant was provided to the condenser/separator from an external supply. Both temperature and flow rate could be controlled-temperature by the supply controller and flow rate by MV12 and MV13. The condensate removed by the condenser/separator was collected in a storage tank (WST1) which could be emptied either automatically or manually.

Various sensors located in the B-CRS provided control and monitor functions. Thermocouples monitored both reactor (inside cartridge) and heater temperature. Two solid-state temperature controllers maintained the constant reactor tempera- ture and protected the heater from excessive duty cycle operation and overheating. Pressure gauges located on the subsystem and TSA instrumentation provided the bulk of the raw data. These are discussed in the following section.

Test Support Accessories

Test Support Accessories were needed for the control, operation and monitoring of the B-CRS. Generally, TSA developed under prior programs was modified as required to fit the needs of the present tests. As an example, Figure 8 shows the B-CRS TSA cabinet containing the gas supply valves, gauges and flowmeters. Only those elements used in the present program relating to the schematic of Figure 7 are indicated.

Feed Gas Control

A major change in operating philosophy for feed gas control was implemented. Under prior testing programs, feed gases were supplied by upstream subsystems in amounts determined by those subsystems. However, this testing program was designed to determine Bosch performance as a function of various operating parameters. Therefore, it was necessary to modify the feed control so that the Bosch subsystem would draw from the H_2/CO_2 mixture supply only those amounts of gases required for its operation. This "on demand" philosophy meant that one parameter had to be selected to be controlled. This parameter was recycle loop pressure at the feed point. The control was implemented with a forward pressure regulator located between the H_2/CO_2 supply and the Bosch feed point. The regulator was set at a nominal 21 kPa (3 psig) level. Whenever the loop pressure was below 21 kPa (3 psig) the feed gases would flow at the maximum. As the loop pressure increased the feed gas flow rate would decrease and eventually cease after the pressure exceeded 21 kPa (3 psig). As a result, a given combination of operating parameters would determine the necessary feed flow rate to operate at maximum performance for that set of conditions. Other conditions would have a different maximum feed flow rate and therefore a different performance. Performance in these tests was always measured as CO_2 reduction rate in kg/unit time.

Coolant Temperature
Indicators and
Shutdown Controllers
(T5 and T6)

Makeup H$_2$ Flow to
B-CRS (FM3)

B-CRS Feed
Gas Pressure (PG11)

H$_2$ Supply
Pressure (PG12)

CO$_2$ Purge To
B-CRS (FM1)

CO$_2$ Flow to
B-CRS (FC2)

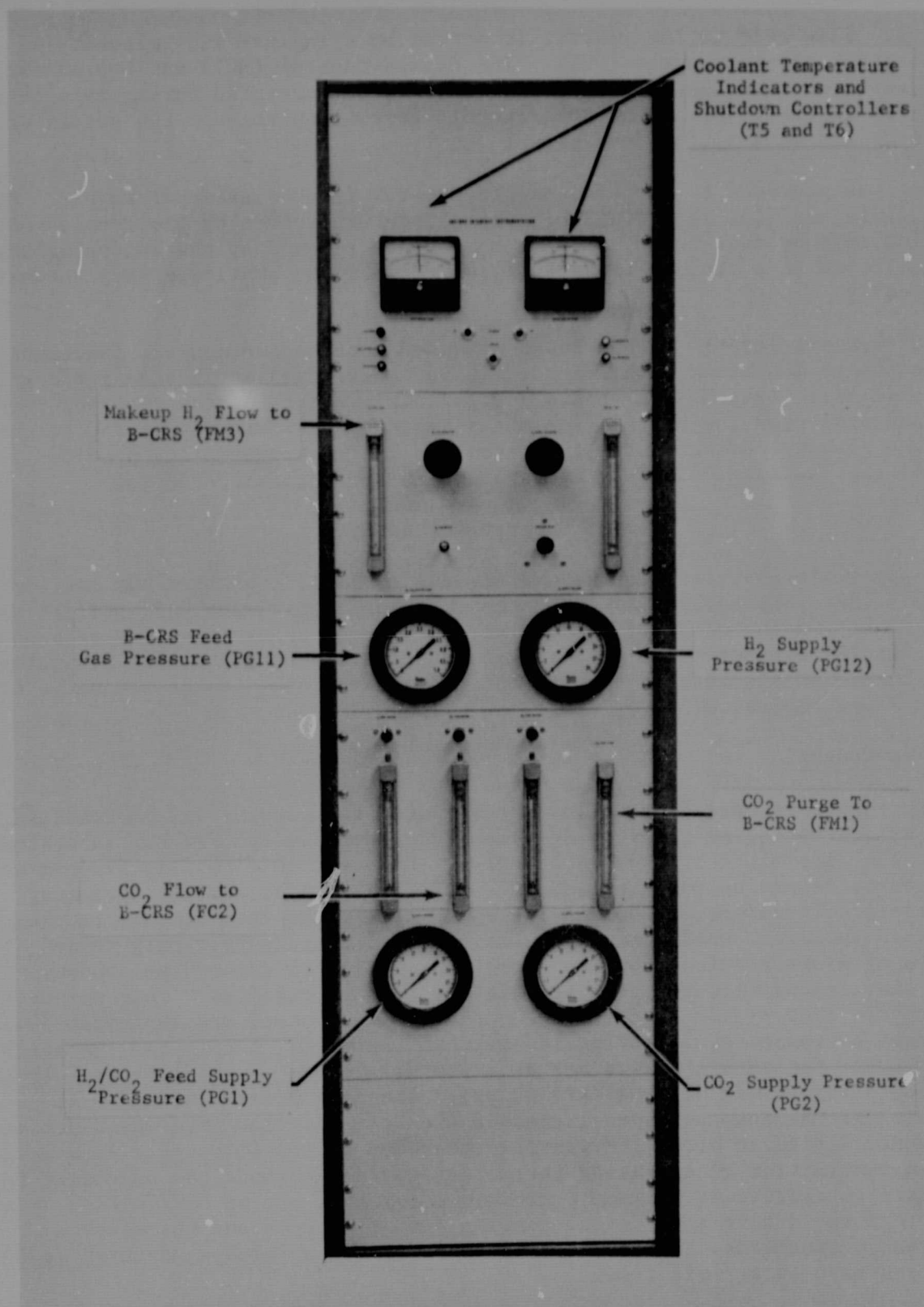H$_2$/CO$_2$ Feed Supply
Pressure (PG1)

CO$_2$ Supply Pressure
(PG2)

FIGURE 8   B-CRS TEST SUPPORT ACCESSORIES CABINET

Water Collection

The measurement of Bosch performance was CO_2 reduction rate. Obtaining instantaneous CO_2 inlet flow rates was difficult. An integrating mass flowmeter would be required since instantaneous readings of flow rate would be questionable because of small perturbations in flow rate. Therefore, water production rate was used for the measurement of performance. This parameter is directly related to CO_2 reduction rate through the overall reaction equation (equation 1).

The water collection hardware described earlier allowed measuring water production rates in cm^3/min over both short and long periods of time. Generally, each sample data point was recorded at hourly intervals or less. The water collected was that produced over the time interval following the preceeding sample point. Measurement error was such that sampling intervals of less than 15 min would give a CO_2 reduction rate within $\pm 10\%$ error. Measurements every half hour or longer gave accuracies within $\pm 3\%$. The measurement was accomplished by placing the water collection hardware in manual mode and draining the water storage tank into a graduated cylinder. This measurement determined the water produced since the prior data point.

Strip Chart Recorders

Five continuous strip chart recorders monitored the Bosch performance from startup (heater application) to shutdown. Four single channel recorders monitored H_2/CO_2 mixture feed flow rate in cm^3/min, feed composition in % CO_2, recycle loop composition in % H_2 and recycle loop pressure in psig. A dual channel recorder monitored the voltage and current supplied to the B-CRS to give an indication of total system power. Information recorded on these recorders was used principally to detect trend changes over long periods of time.

Coolant Control

Both the flow rate and temperature of the inlet coolant could be maintained by a modification of the coolant control hardware supplying the B-CRS. Two hand valves and a flowmeter were installed to control the flow to the condenser/separator from a constant flow coolant supply. The temperature of the coolant to the inlet of the condenser/separator could be controlled by a setting on the coolant supply.

During the actual testing it was desired to vary the dew point of the recycle loop downstream of the condenser/separator. The most efficient way of doing this was by changing the condenser/separator inlet coolant temperature. Changing the coolant flow rate through the condenser/ separator would also have the same effect, but this effect is masked somewhat by the design of the condenser/separator as a heat exchanger. To rule out this design influence, the coolant flow rate was adjusted to give a sufficient flow rate through the condenser/separator to maintain a small temperature differential (outlet to inlet) across the condenser/separator. In this manner, the temperature of the coolant exiting the condenser/separator was assumed to approximate the recycle gas dew point temperature.

PROGRAM TESTING

The test program was designed to measure the response in performance of the B-CRS as a function of changes in five operating parameters. These parameters were: reactor operating temperature, recycle loop composition (% H_2), recycle loop dew point, catalyst weight and recycle loop flow rate. A total of 12 reactor cartridges were used. Generally several levels of one parameter were achieved for each cartridge. Each cartridge was operated for an average of 11 hours. All the tests were successfully completed. The test durations and the number of data points taken met or exceeded test program goals.

Test Procedures

The overall approach of the test program was to vary one parameter at a time while maintaining the other parameters at nominal values. The nominal values were somewhat arbitrary but were based on past operating experience. In a complex chemical reaction system, like the Bosch, changing several parameters at one time could influence the optimization of other parameters in terms of overall subsystem performance. Ideally, a rigorous testing program would evaluate the effect of all possible combinations of parameters. If only three different levels of each parameter is considered a minimum to generate a reasonable performance curve, a total of 243 different experiments would be required for evaluating five parameters. This number far exceeded the test time and funding allocated. Therefore, the approach taken was to vary one parameter while holding the others constant. Table 2 shows the nominal value and range of the parameters covered. Actually, instead of three data points, most parameters were varied to five or six different levels within the range.

For each test a fresh cartridge was installed into reactor 1 of the B-CRS and the subsystem was then started. The order of varied parameters was randomly selected to minimize any systematic error in startup procedure or data taking. Nominal operating conditions of the B-CRS are shown in Table 3. This table includes the nominal operating conditions of the five principle parameters. Following heater power application, normally one to two hours is required for the reaction to be initiated and the subsystem to reach steady-state.

The general procedure for sample data taking was the following. After establishing an assumed steady-state, a data point was taken. After waiting another half hour to an hour (depending on the variable) a second data point was taken. A short time later a third point was taken. If the CO_2 reduction rate, as determined by water collected over the intervening time periods, remained constant, then the next level of the parameter under study was selected. In other words, at least two consecutive data points giving the same CO_2 reduction efficiency (within the experimental error) were required before the parameter was changed to a new level. Experiments performed in this manner allowed completion of a test within 11 to 15 hours and sufficient data taken to plot a curve. As mentioned previously, the cartridges are nominally sized for 12 person-day operation. At an assumed maximum CO_2 reduction rate equivalent to a five person-level, approximately 60 hours of operation could be expected. Completing a test within 15 hours allowed operating time to be ruled out as an influencing parameter in the experiment design.

TABLE 2   PARAMETER RANGE

		Low	Nominal	High
1.	Temperature, K (F)	811 (1000)	933 (1220)	977 (1300)
2.	Loop Composition, % H_2	36	45	52
3.	Recycle Flow Rate, g/min (lb/h)	52 (6.9)	65 (8.6)	70 (9.3)
4.	Coolant Flow/Temp. Flow, dm^3/min Temp., K (F) (inlet)	5 275 (35)	5 276 (38)	5 294 (70)
5.	Catalyst Weight, g (lb)	80 (0.18)	150 (0.33)	250 (0.55)

TABLE 3   NOMINAL TEST OPERATING CONDITIONS

Heater Control Temperature, K (F)	1073 (1473)
Reactor Temperature, K (F)	933 (1220)
Recycle Flow Rate, dm^3/min (cfm)	65 (2.3)
Coolant Flow Rate, dm^3/min (lb/min)	5 (11)
Coolant Inlet Temperature, K (F)	276 (38)
H_2/CO_2 Feed Mixture	
Flow Rate	Variable (On Demand)
Source Pressure, kPa (psia)	116 (16.9)
Feed Pressure, kPa (psia)	99.3 (14.4)
Composition, % CO_2	40
Reactor Inlet Pressure, kPa (psia)	145 (21)
Ambient Pressure, kPa (mm Hg)	97.4 (14.1)
Ambient Temperature, K (F)	296 (74)
H_2 Makeup	
Flow Rate, dm^3/min (cfm)	1.1 (3.9×10^{-2})
Pressure, kPa (psia)	121 (17.6)

Parametric Tests

The test results on the effects of five operating parameters are presented
below. The order of presentation was selected in terms of decreasing order of
parameter importance on overall B-CRS operation.

Reactor Temperature

Reaction temperature appears to be the most important operating parameter,
because it directly relates to the reaction constant of equation 1. The Bosch
reaction actually occurs within a wide temperature range. During the initial
startup from ambient temperature, the reaction was consistently found to start
at temperatures as low as 533 K (500 F). The reaction rate at this temperature
is low but increases with increasing reactor temperature.

Subsystem performance as a function of reactor temperature is shown in Figure 9.
This curve shows that a maximum level of approximately 5.6 kg/d (12.3 lb/d) of
CO_2 can be reduced with the particular B-CRS hardware utilized. The maximum
temperature occurred at approximately 930 K (1215 F). Reactor performance
decreased for temperatures above this value. It should be recalled that each
data point shown in the figure represents an average of several samples taken
before the parameter was varied to the next level. Experimental procedure
also established that the temperature levels were not always taken in order.
In other words, first a low value could be taken, then a higher value, then a
lower value, etc. This was done to minimize experimental error.

Results shown in Figure 9 agreed with projections. The shape of the curve
also indicates that a decrease in reactor operating temperature from 930 K
(1215 F) of approximately 100 K (180 F) would cause only a 20% reduction in
performance. Operating at a lower temperature may be desirable in some cases
where heater power needs to be conserved during periods which coincide with
reduced CO_2 reduction requirements, e.g., spacecraft dark cycle operation.

Recycle Loop Composition

The composition of gases in the recycle loop is a mixture of CO_2, CO, CH_4, H_2,
CH_4 and water vapor. Of these, only CO_2 and H_2 are controllable on a practical
basis, since they are the only gases that enter the subsystem from outside
sources. The reaction stoichiometry shows that 67% of the incoming gases must
be H_2. However, the percent H_2 in the recycle loop can vary. The instrumenta-
tion employed in the B-CRS to control the stoichiometry of the incoming gases
was based on measuring and controlling the volumetric concentration of H_2 in
the recycle loop. Therefore, the percent H_2 in the loop composition was
varied instead of percent CO_2.

Figure 10 shows the results of the loop composition tests. The S-shaped curve
shows that greater than 45% H_2 is desired to obtain optimum performance. As
the H_2 percentage in the recycle loop increases, the percentages of other
gases also vary and generally decrease. The richer H_2 mixture influences not
only the primary equation (1) but intermediate equations, in particular the
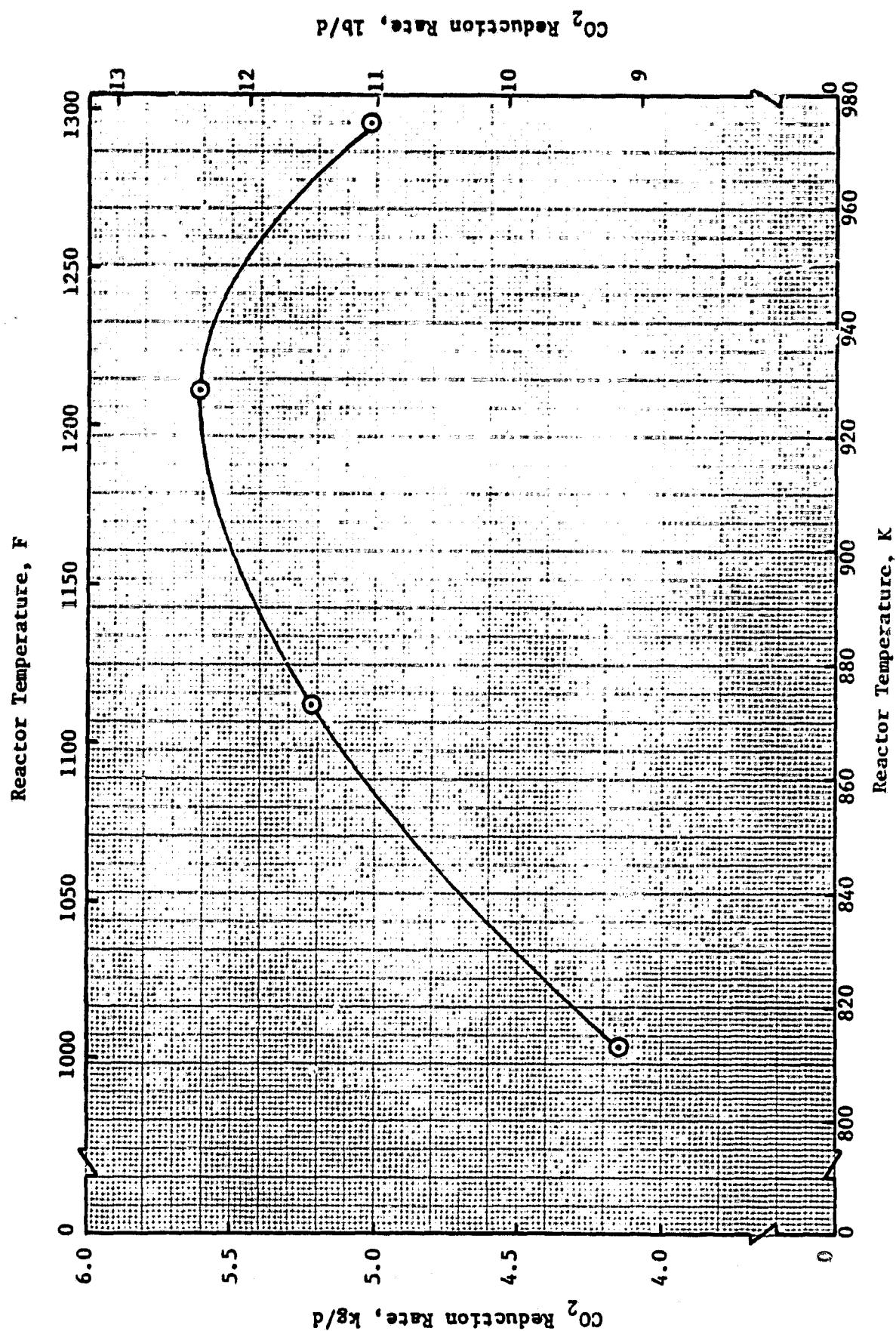water-gas shift equation (4).

FIGURE 9   B-CRS PERFORMANCE AS A FUNCTION OF REACTOR TEMPERATURE
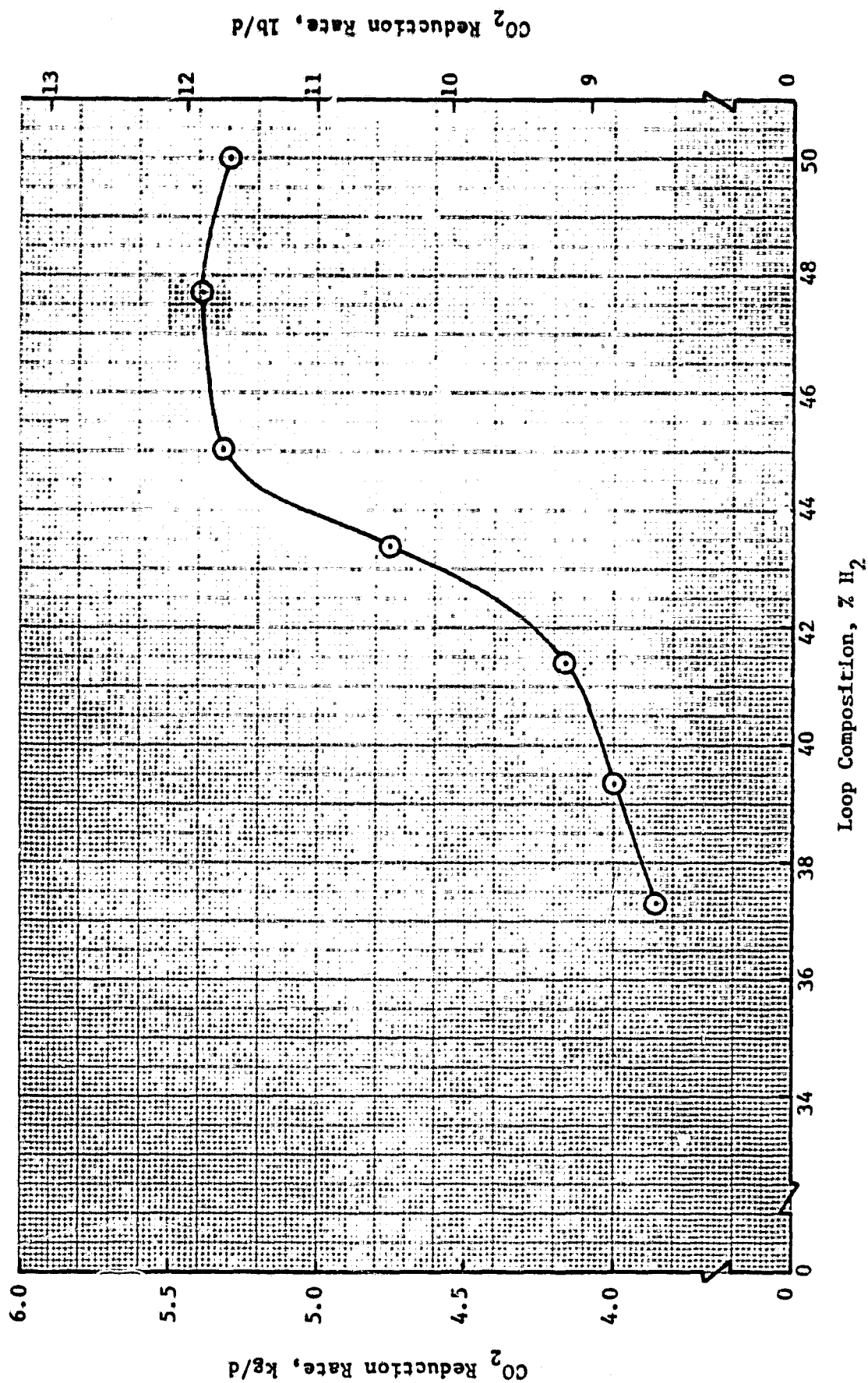
FIGURE 10 PERFORMANCE AS A FUNCTION OF RECYCLE LOOP COMPOSITION

Recycle Loop Dew Point

The recycle loop dew point was adjusted by controlling the temperature of the coolant to the condenser/separator. A large absolute coolant flow rate through the condenser/separator was maintained throughout the testing to ensure that the outlet coolant temperature would be close to the inlet coolant temperature. In this way the recycle loop dew point temperature was maintained very nearly equal to the outlet coolant temperature.

Figure 11 shows the B-CRS performance response to dew point variations. The data shows that higher water vapor levels in the recycle loop severely impact performance. The curve also indicates that the lower coolant temperatures are desirable. However, the B-CRS may have to accept coolant at available temperatures in many spacecraft applications. This could be as high as 294 K (70 F). When this is the case, the design of the condenser/separator would be an important consideration. Heat transfer rate, surface area and condenser/separator configuration would be the major design factors to maintain the dew point at reasonably low levels.

Recycle Flow Rate

Recycle flow rate influences performance, because it determines the amount of reactants exposed to reaction sites per unit time and space velocity and residence time within the cartridge. As in most flow reactors increasing flow rate will increase conversion efficiency. This was found to be true in the B-CRS, as shown in Figure 12. Optimum performance occurred at approximately 64.5 g/min (8.5 lb/h). For the corresponding feed rate at this value, the ratio of recycle mass flow rate to feed mass flow rate was approximately 15 to 1.

Recycle flow rate is an important factor in sizing the compressor. For minimum subsystem power and volume, a small compressor is desired. The data obtained indicates that for an efficient Bosch design at the four to six-person level the compressor must handle at least 4.5 kg/h (10 lb/h).

Catalyst Density

Prior experience of Life Systems and that of other investigators had established a nominal catalyst charge of approximately 12 g/person-day (0.026 lb/person-day). Consequently, a nominal value of 150 g (0.33 lb) of pretreated catalyst was installed in each cartridge. This amount corresponds to a density of 25 g/dm^3 (1.56 lb/ft^3). The objective of this test was to examine the effect of changing catalyst density on performance. Figure 13 shows the influence of catalyst density over a range of 10 to 45 g/dm^3 (0.62 to 2.8 lb/ft^3). Performance rapidly drops off if less than 30 g/dm^3 (1.87 lb/ft^3) is used and a catalyst density of greater than this amount appears not to improve performance significantly.

Figure 14 shows the same data as Figure 13, but plotted as a function of time for the various catalyst densities. During the first three hours of operation, no data points were taken as the reactor reached initial operating temperature and the reaction rate stabilized. The performance curves for the two highest
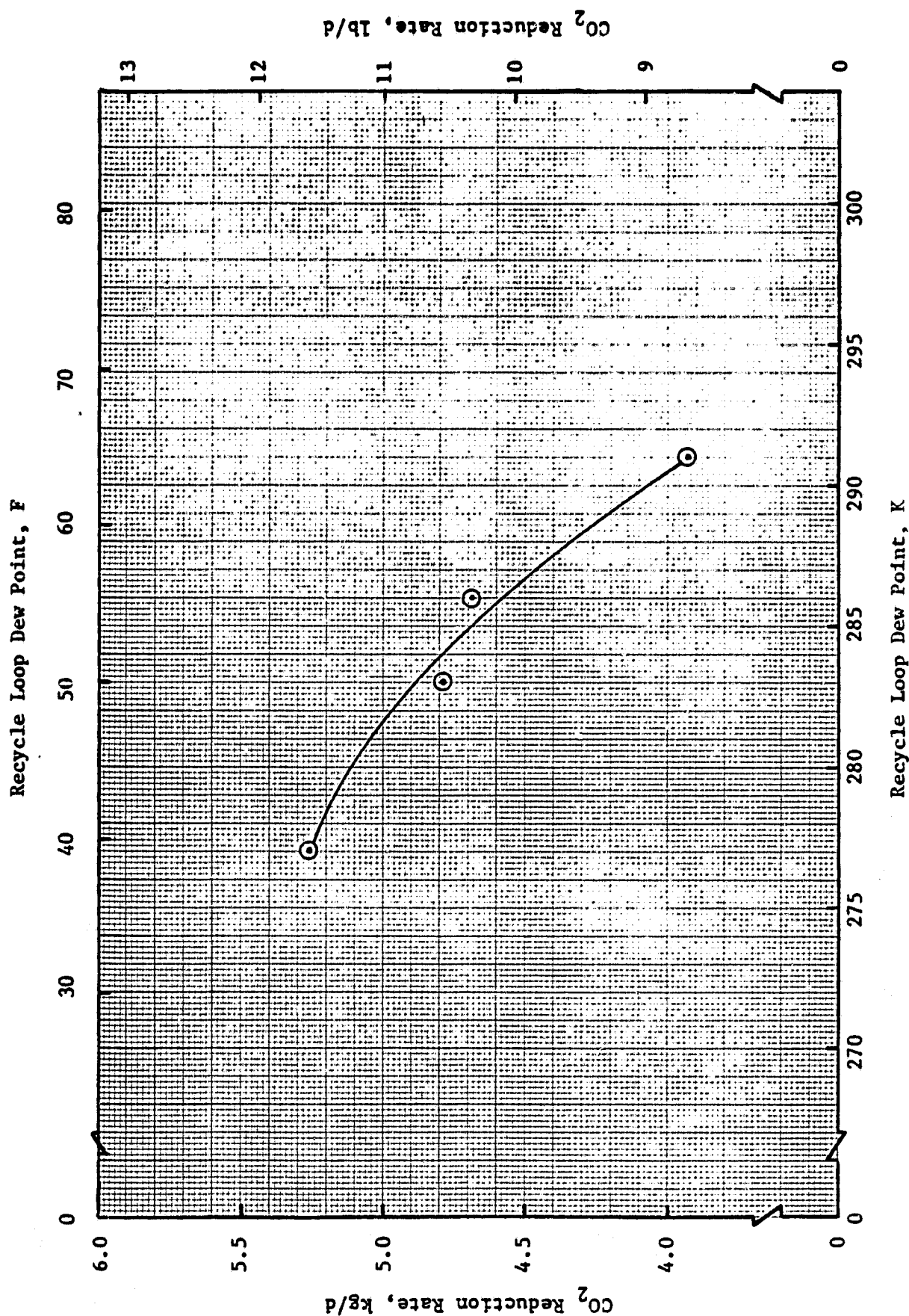
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FIGURE 11   PERFORMANCE AS A FUNCTION OF RECYCLE LOOP DEW POINT
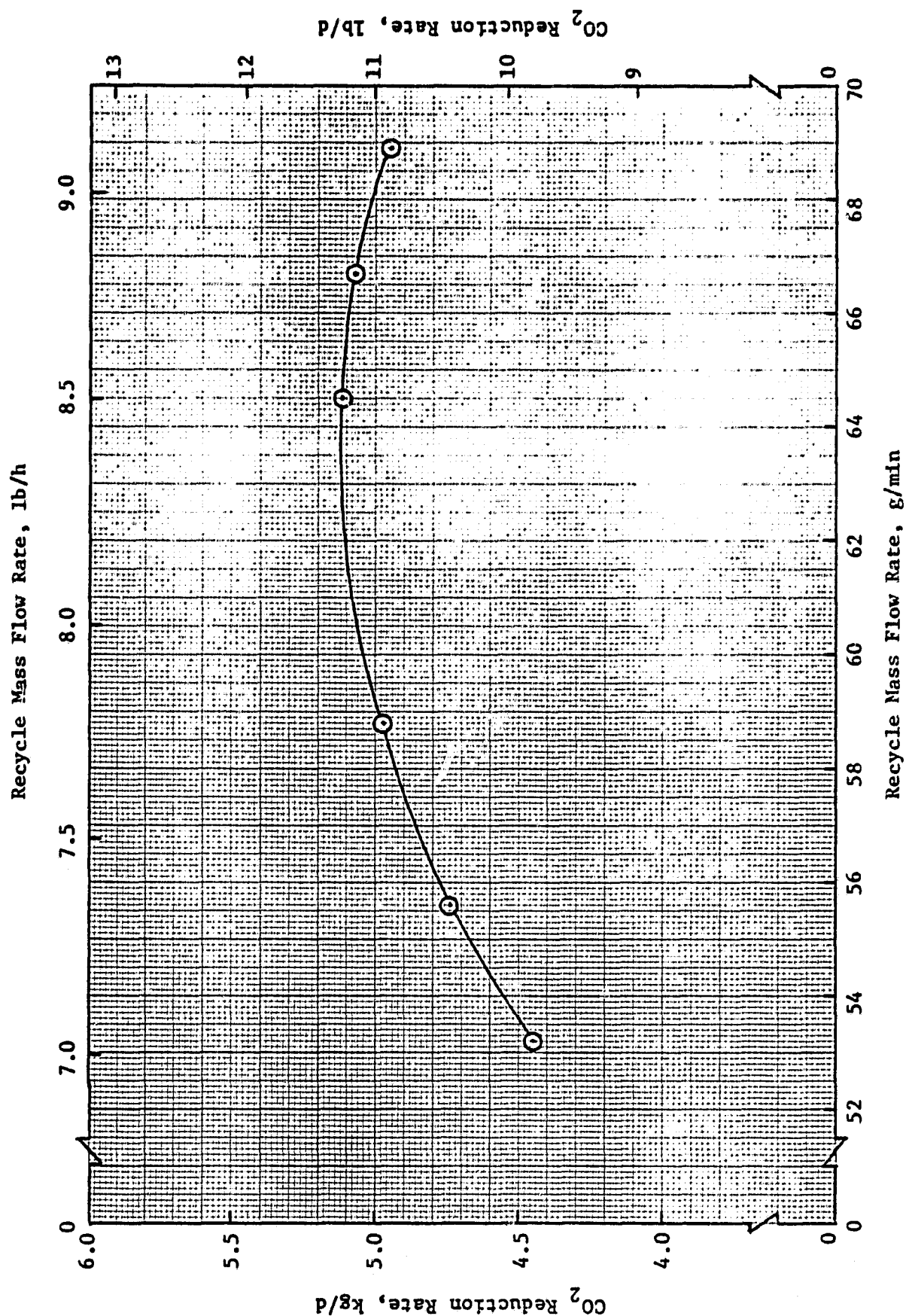
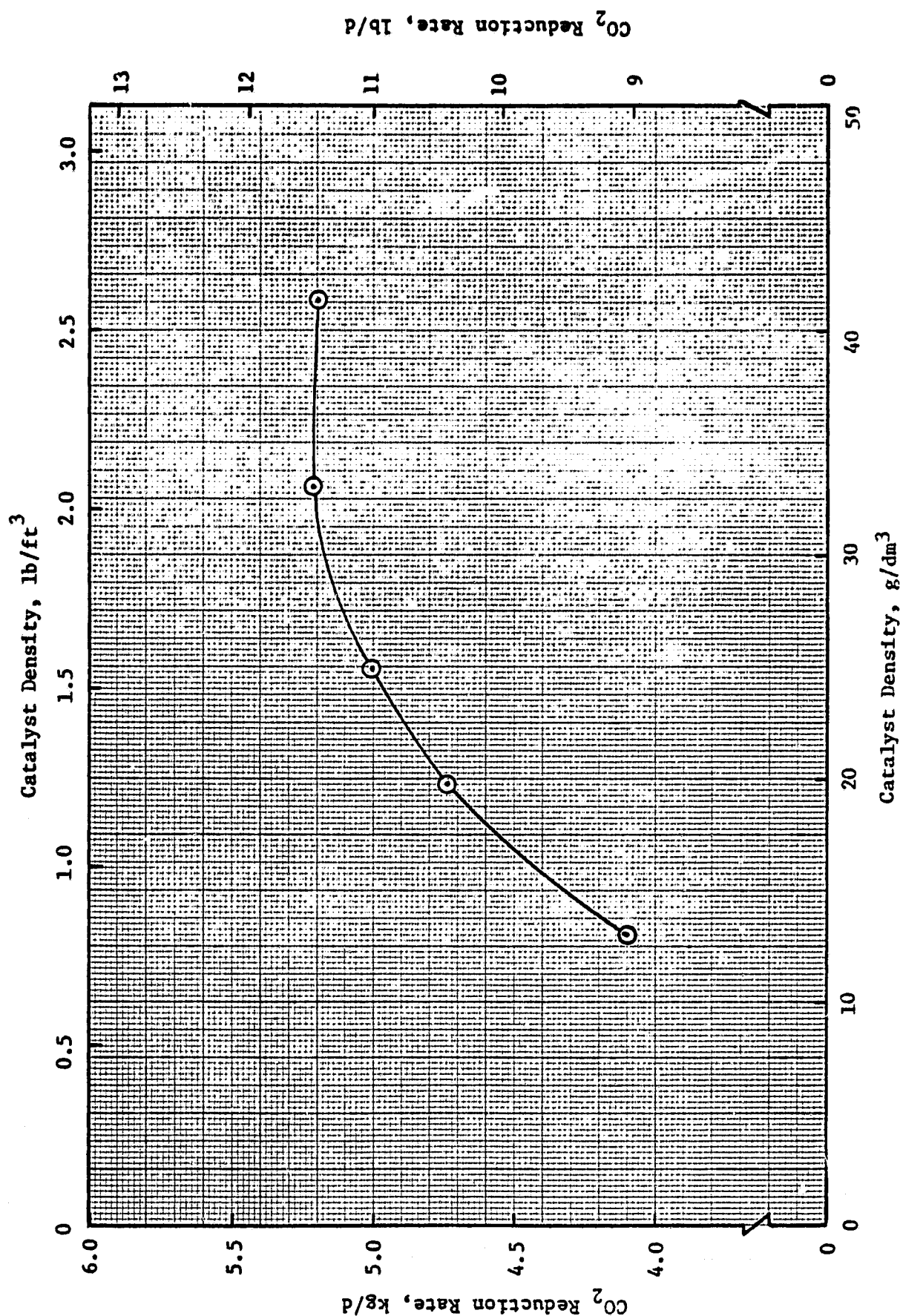FIGURE 12 PERFORMANCE AS A FUNCTION OF RECYCLE FLOW RATE

Life Systems, Inc.

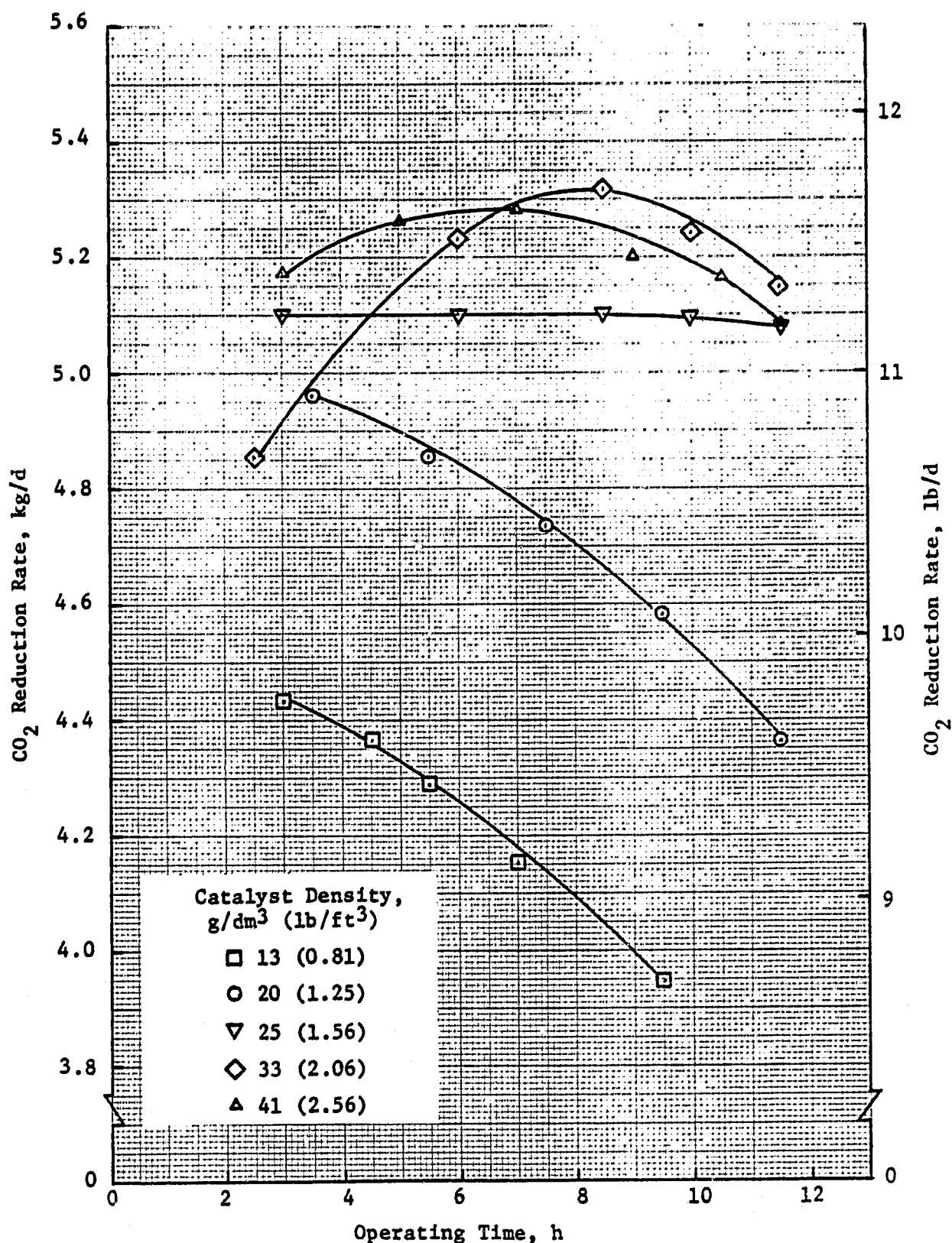FIGURE 13   PERFORMANCE AS A FUNCTION OF CATALYST DENSITY

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FIGURE 14   PERFORMANCE AS A FUNCTION OF
OPERATING TIME FOR VARIOUS CATALYST DENSITIES

catalyst loadings show deviations from the expected initial maximum performance. These curves show that maximum performance was not reached until approximately 6 to 8 h into the runs. The delayed optimum performance could be due to nonoptimum levels of other parameters. These results are not clearly explained by the data or theory. The more typical behavior of a cartridge is as shown in the other three curves. Reactor performance generally is best within the first third to a half of cartridge capacity and eventually will fall off with time. The nominal catalyst loading, i.e., 25 g/dm^3 (1.56 lb/ft^3) showed the best sustained performance.

Figure 15 gives a slightly different measure of B-CRS performance over time. Here the ordinate is mass of carbon produced expressed as an amount instead of a rate. Of course, water produced, H_2 consumed or CO_2 reduced could have been selected as well. The data points for three catalyst loadings are shown along with calculated curves representing constant one to five person level production. Similar values could also have been obtained by integrating over time the curves of Figure 14. The comparison with the person-level curves indicates that although rate performance changes over time, a given catalyst density can support only one maximum feed rate (corresponding to a given person-level) over extended periods of time.

Reactor startup time is an important parameter, because it relates to the time required for switchover from an operating cartridge to a dormant cartridge for continuous CO_2 reduction capability. Table 4 shows that reaction startup time was directly proportional to catalyst density. This could be explained from fundamental reasoning. The carbon collection cartridge is mostly void space, initially containing only the catalyst. The catalyst (steel wool) has a certain heat capacity. The more steel wool the more heat that is retained. Also additional steel wool tends to uniformly distribute the heat faster so it can be expected that the cartridge will heat up more uniformly in a shorter period of time. If power supplied by the heater at the central axis of the cartridge can be distributed to the outside in a shorter period of time the cartridge heatup from ambient to reaction initiation temperature is much shorter. This was verified by the temperature measurements.

Testing Summary

Tables 5 and 6 summarize the test results obtained with the 12 cartridges. The data was consistent and no anomalies were found, even though the tests were conducted with one particular set of B-CRS hardware. This hardware was developed approximately 10 years ago and has accumulated over 10,000 operating hours, a testimonial to Bosch technology readiness. The design of the experiments was carefully selected to rule out or minimize influences of subsystem, TSA or component hardware factors. The results of the experiments are applicable to future B-CRS hardware.

CONCLUSIONS

Based on the results of this program, the following conclusions are drawn:

1. The relative importance of the five operating parameters investigated is, (in decreasing order): reactor temperature, percent H_2 in the recycle loop, recycle loop dew point downstream of the condenser/separator, recycle flow rate and catalyst density. Other paramters (e.g., gas
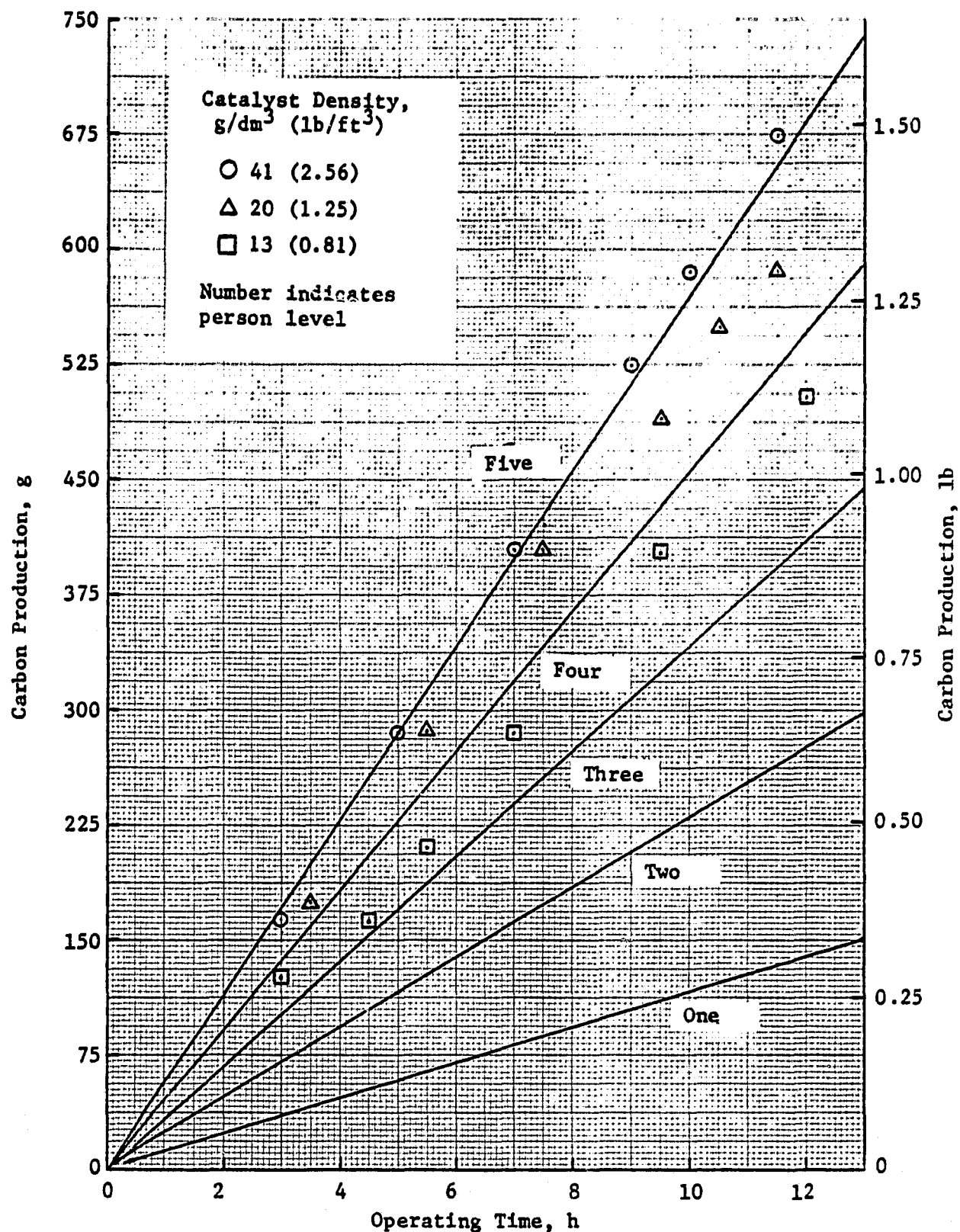
FIGURE 15   CARBON PRODUCTION VERSUS OPERATING TIME

TABLE 4 REACTOR STARTUP TIME

Catalyst Density, g/dm^3 (lb/ft^3)	Nominal Startup Time, h[a]
13 (0.81)	1.50
20 (1.25)	1.33
25 (1.56)	1.25[b]
30 (1.87)	0.90
33 (2.06)	0.74
41 (2.56)	0.67

(a) Defined as time interval from initial heater application
until reactor accepts additional reactants.
(b) Average value of four cartridges.

TABLE 5   CARTRIDGE DATA RESULTS

No.	Catalyst Density, g/dm^3	Total Water Collected, kg (lb)	Total Carbon Collected, kg (lb)	Operating Time, h
1	25	4.62 (10.20)	1.240 (2.73)	31.0
2	25	1.83 (4.04)	0.627 (1.38)	11.4
3	25	5.49 (12.10)	1.870 (4.12)	35.3
4	25	2.18 (4.81)	0.718 (1.58)	13.0
5	25	1.81 (3.99)	0.618 (1.36)	10.2
6	20	1.80 (3.97)	0.611 (1.35)	11.5
7	41	2.25 (4.96)	0.803 (1.77)	12.8
8	25	2.23 (4.92	0.745 (1.64)	12.7
9	25	1.88 (4.15)	0.654 (1.44)	11.5
10	33	2.29 (5.05)	0.778 (1.72)	13.0
11	25	2.18 (4.81)	0.734 (1.62)	13.5
12	13	1.95 (4.30)	0.655 (1.44)	14.5

TABLE 6   BOSCH CARTRIDGE SUMMARY

No.	Parameter Varied	Total Operating Time, h	Overall CO_2 Reduction Rate[a] kg/d (lb/d)
1	Reactor Temperature	31.0	3.92 (8.64)
2	Reactor Temperature	11.4	4.77 (10.50)
3	Loop Composition	35.3	4.61 (10.20)
4	Reactor Flow Rate	13.0	4.96 (10.90)
5	Reactor Flow Rate	10.2	5.28 (11.60)
6	Catalyst Weight	11.5	4.63 (10.20)
7	Catalyst Weight	12.8	5.36 (11.80)
8	Reactor Flow Rate	12.7	5.13 (11.30)
9	Normal Operation	11.5	4.90 (10.80)
10	Catalyst Weight	13.0	5.21 (11.50)
11	Coolant Temperature	13.5	4.76 (10.50)
12	Catalyst Weight	14.5	3.96 (8.73)
	Total	190.4	

(a) Average based on total water and carbon collected.

distribution in the cartridge) may be important but the above five are concluded to have the most significant effect on reactor performance and, therefore, reactor size.

2. A Bosch reactor should operate at 922 ±83 K (1200 F ±150 F). The reactor design should account for heat losses by conduction, convection and radiation at this nominal operating temperature. Reactor design should include heat of evolution (function of person-size) so that the net heater power required can approach zero.

3. The control technique of sensing recycle loop composition H_2 percentage and adding makeup H_2 to bring the total feed flow rate to the stoichiometric ratio ($H_2:CO_2 = 2:1$) has been proven to be a most effective control approach. This technique is particularly amenable to integrating the B-CRS with an EDC.

4. Performance of the B-CRS drops off significantly as a function of recycle loop dew point. This fact should be taken into account when sizing and designing the condenser/separator and selecting spacecraft coolant availability and temperature. As low a temperature as possible (above freezing) is desired.

5. A minimum catalyst density of 30 g/dm^3 (1.87 lb/ft^3) is required for good performance. Values over this quantity will not change performance markedly but will decrease reactor startup time. However, since the catalyst is expendable, logistics will increase. Because of the negligible cost of the present catalyst, using slightly more than the minimum amount is proper. Therefore, catalyst density in the range of 30 to 35 g/dm^3 (1.87 to 2.18 lb/ft^3) is recommended. At the latter density, the total catalyst required for a three-person, 100 day mission is only 5.3 kg (11.7 lb).

RECOMMENDATIONS

Based on the results of this study, it is recommended that further work be performed to: Design, fabricate, assemble and test a preprototype B-CRS for NASA's Regenerative Life Support System Evaluation (RLSE) Program. In that program a Sabatier CO_2 reduction unit has already been built and is presently undergoing tests. Development of a B-CRS to the same requirements as the Sabatier unit and implementation into the RLSE would close the development status gap between the two approaches.

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